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DISTRIBUTION OF ENVIRONMENTAL PLUTONIUM IN THE TRINITY SITE ECOSYSTEM AFTER 27 YEARS

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Abstract

The results are presented for a radioecological survey of the Trinity Site environs, where the world's first (July 1945) atomic bomb was detonated. The temporal behavior of the low environmental levels of the plutonium produced by this detonation are discussed. The data from this study were compared with similar data obtained in the Trinity Site environs nearly 20 years ago. The major change which was observed was an increased migration of Pu into the soils. Concentrations of Pu in vegetation and rodents were too low to make valid comparisons.

Introduction

An ecological investigation of plutonium was initiated in the fallout pathway of Trinity, the first nuclear detonation, which occurred on July 16, 1945 in southern New Mexico. Trinity Site was especially interesting as a study area because of the "aged" nature of the radioactive debris distributed in the area. In addition, it was of interest to gather ecological data on plutonium in the xeric Trinity Site environs to compare with similar data being gathered in several semi-mesic ecosystems at the Los Alamos Scientific Laboratory in northern New Mexico.[1]

The data presented in this paper were obtained from samples gathered during one sampling period in the Trinity Site environs on September 27-28, 1972. The primary objectives of this effort were to survey the plutonium content of a few ecosystem components and to measure the field gamma radiation intensity as a function of distance from Ground Zero (GZ) some 27 years after the detonation to facilitate the design of more intensive studies.

Methods and Materials

Trinity Site, a fenced area immediately around GZ, is located in the semi-arid northern portion of the Tularosa Basin about 40 miles SW of Socorro, New Mexico, on the White Sands Missile Range (Fig. 1). A general description of the physiography, climate, vegetation and mammals of the area around Trinity Site can be found in various references.[2,3,4]

A transect was established along the reported fallout pathway of the nuclear debris from the detonation (Fig. 1), utilizing maps constructed by University of California scientists in 1948.[5] Nine sampling locations were established on the transect, two in the GZ area, and the remainder at 8 km increments to a distance of about 56 km from GZ.

One soil core was taken at each location with a disposable 30 cm section of 2.4 cm diameter polyvinyl chloride pipe. The pipe and contained core from

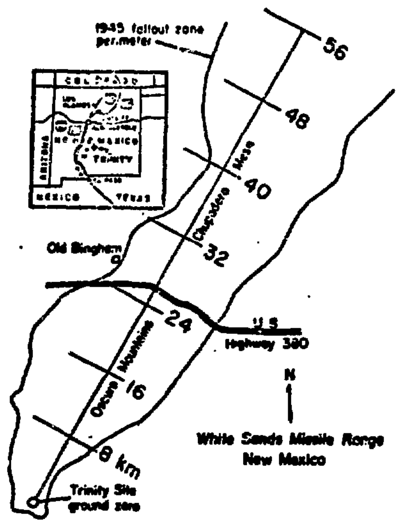

Fig. 1. Sampling transect utilized for the radioecological resurvey of Trinity.

each station was sealed in a plastic bag, frozen on return to the laboratory and sectioned into a 0-2.5 cm, 2.5-7.5 cm and 7.5-30 cm segment.

A sample of the most abundant forb, grass, and shrub/tree species was collected where possible and was individually sealed in plastic bags. Samples included the above-ground portions of the grasses and forbs and the terminal leaves and stems of the shrub/tree species. Dust on the plant surfaces was not removed prior to analysis.

Rodents were collected with peanut butter baited snap traps and were bagged and frozen for later dissection. Tissues analyzed for plutonium included lungs, liver, hide, and carcass (skeleton and skeletal muscle). Care was taken during the dissection to avoid cross-contaminating the soft tissues with hair from the pelt.

Trinitite, the fused soil material formed by the intense heat produced by the detonation, was also collected to determine its plutonium content and to identify the gamma emitters present.

Analytical procedures for plutonium included a combination of wet-dry ashing techniques utilizing a muffle furnace and HNO_3-HF solutions followed by ion exchange column separation of plutonium, electrodeposition and alpha ray spectroscopy for both ^{238}Pu and ^{239}Pu. The minimum sensitivity of the alpha counting system based upon background counts during a 24 hour period was 0.03 pCi ^{238}Pu or ^{239}Pu/sample ($\alpha = 0.05$).

All soil and vegetation samples contained sufficient Pu and/or were of sufficient mass to reduce the relative counting standard deviation on each sample to less than 25 percent ($1\ \sigma$). However, the generally low Pu content and small mass of certain rodent tissues resulted in relative counting standard deviations of as much as 100 percent. The standard errors associated with the rodent data presented later, reflect this fact.

Direct measurement of radiation in the field was accomplished with a Ludlum Model 125 Count Rate Meter, which utilizes a NaI (Tl) scintillation detector, for *in situ* environmental gamma radiation measurements. This instrument's readout was calibrated to give a proper "μR/h" reading with ^{60}Co gamma rays. During the survey, the instrument was held at about 3 feet above the ground surface and the observed rate noted and recorded at the respective locations. Because of an inherent photon energy-dependent response, all readings obtained with the Ludlum were normalized using 10 Los Alamos Scientific Laboratory environmental radiation dosimetry stations which utilize LiF thermoluminescent dosimeters (TLD) for background radiation measurements.[6] Dosimetric values obtained from these TLD materials have been shown to be essentially independent of radiation energy and, therefore, provided a basis for the correction or normalization of the Ludlum Model 125 meter readings. The normalization assumed uniform photon spectral distribution. The observed average ratio of TLD-determined exposure rates to the survey meter measurements was 0.70.

Results and Discussion

The Pu content (^{239}Pu and ^{238}Pu) of all sample types as a function of distance from GZ is presented in Table 1.

Table 1. The $^{238-239}Pu$ content of some ecosystem components collected in the fallout zone of the Trinity detonation.

	Kilometers from Ground Zero (GZ) Along Fallout Pathway								
	GZ	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
Soils (fCi/g dry)									
0-2.5 cm	254812	36	434	312	123	399	600	270	1441
2.5-7.5 cm	262730	36	0	226	746	116	344	20	89
7.5-30 cm	61858	23	6.6	213	173	3.4	62	30	36
Vegetation (fCi/g wet)*									
Grasses	740	140	73	36	3.1	15	19	11	30
Forbs				3.0	0.60	2.5	4.7	3.6	16
Shrubs/trees	6.2	2.4	9.2		1.9	4.1	6.5	0.60	8.3
Rodents (fCi/g wet)*									
Liver		12 (11)**	4.4 (2.7)	1.3 (0.9)	1.7	7.3 (1.3)	4.5 (2.7)	6.2	
Lungs		25 (19)	11 (8.1)	4.1 (3.9)	3.3	40 (27)	50 (63)	0	
Hide		4.8 (0.96)	5.6 (2.3)	2.0 (2.0)	0.75	3.4 (1.7)	2.0 (1.1)	3.0	
Carcass***		2.2 (1.2)	13 (8.0)	4.2 (1.9)	0.06	30 (29)	4.7 (1.6)	2.5	
No. of samples		4	3	3	1	5	4	1	

* Species comprising the vegetation and rodent samples are given in the text.

** Parenthetic value represents the standard error of the determination.

$$\text{Standard Error} = \frac{\text{standard deviation}}{\sqrt{\text{No. of samples}}}$$

*** Carcass includes skeleton and skeletal muscle.

The Pu data for vegetation (fCi/g wet) were summarized according to the type of plant (grass, forb or shrub/tree) to provide some basis for viewing Pu concentration gradients with distances from GZ since none of the plants were found at every sampling station. Grass species included *Tridens pulchellus*, *Sporobolus Neallei* and *Bouteloua eriopoda* while the forb category included *Mirabilis multiflora*, *Conyza Coulteri*, *Dithyrea Wislizeni*, *Aphanostephus humilis* and *Melilotus albus*. All the grass and forb species were generally less than 60 cm tall; the shrub/tree species which included *Atriplex canescens*, *Larrea tridentata*, *Lycium Andersonii*, *Juniperus monosperma* and *Rhus macrophylla* were generally greater than 60 cm tall.

The Pu data for rodent tissues (fCi/g wet) were also summarized without regard to species because the inadequate number of samples did not permit a species comparison and because the species composition of the catch changed with distance from GZ. Species caught included *Perognathus flavus*, *Citellus spilosoma*, *Peromyscus maniculatus*, *P. truei*, *Onychomys leucogaster*, *Neotoma mexicana* and *Dipodomys ordi*.

The Pu concentrations in many of the soil core segments (Table 1) were significantly above background. Levels in GZ soils were as much as 10^4 times higher than the 10-100 fCi Pu/g which has been reported for several New Mexico area soils.[7] A maximum of about 260,000 fCi Pu/g was observed at GZ in both the 0-2.5 cm and the 2.5-7.5 cm core segments. The maximum concentration in non-GZ soil (1442 fCi/g) was measured in the 0-2.5 cm segment from the 56.4 km sampling station.

The Pu data for soils from the GZ and 0.1 km stations cannot be compared with the data for the remainder of the transect because the area around GZ was mechanically disturbed shortly after the detonation in an effort to reduce surface radionuclide contamination. Ground Zero, for example, was covered with at least 15 cm of uncontaminated soil and the area around the 0.1 km station was scraped to remove the Trinitite lying on the ground surface. The high Pu concentration in the 0-2.5 cm segment of the GZ soil sample (Table 1) indicates either 1) the overburdening was not successful; 2) that the covered Pu had migrated to the soil surface; or 3) that the overburden had blown away over the last 27 years, thus exposing the contaminated soil.

The Pu concentrations in the 0-2.5 cm segment generally increased toward the distal end of the sampling transect and reached a maximum at the 56.4 km station. Olafson et al.[8], during efforts to map the fallout zone from Trinity also noted that the highest Pu concentration in soils, vegetation and small mammals outside the GZ area occurred about 45 km from GZ.

The vertical distribution of Pu was relatively uniform in most of the core samples from GZ to the 24.1 km station. This suggested that Pu which was initially deposited on the soil surface as much as 27 years ago had migrated at least 30 cm into the soil profile. On the other hand, the Pu in soils from distances greater than 24.1 km was increasingly concentrated in the upper 2.5 cm. Olafson et al.[8] and Olafson and Larson[9] reported that the Pu in Trinity area soils about 20 years ago was almost exclusively confined to the top 2.5 cm of soil.

Many factors could account for a difference in the rate of vertical migration of Pu in soils, including differences in the chemical and physical form of the Pu and/or differences in the chemical, physical, and biological makeup of the environment.[10]

The Pu concentrations in grasses were consistently elevated with respect to similar measurements in other areas of New Mexico.[1,9] On the other hand, the Pu concentrations in forb and shrub/tree samples were generally indistinguishable from worldwide fallout Pu levels in New Mexico vegetation which measure about 1-5 fCi/g wet sample.[1,7] The Pu data for grasses as a function of distance from GZ generally followed the pattern which was observed for the 0-2.5 cm soil core segment (Table 1). The maximum Pu concentration in grasses (768 fCi/g wet) was observed at GZ and decreased to a minimum of 5.1 fCi/g at the 24.1 km station. Pu concentrations in grasses then generally increased with distance.

The Pu data for rodent tissues in Table 1 show a considerable variability. Sources of this variability would include among other things, species differences, the low Pu content of the tissues yielding generally poor counting statistics, and an insufficient number of samples.

In general, rodent lungs had the highest mean Pu concentrations and exhibited a pattern with distance from GZ that was similar to the 0-2.5 cm layer of soil and the grass. Lung deposition of Pu suggested that resuspension of soil may be an important mechanism in the biological radistribution of Pu. Other investigators have noted high lung concentrations in small free-roaming mammals.[11,12]

The activity ratios $^{239}Pu/^{238}Pu$ for all sample types are summarized in Table 2. The ratios for the 0-2.5 cm and 2.5-7.5 cm core segments averaged 19 and 18, respectively, while the 7.5-30 cm segment averaged 9. The mean values for vegetation were about 8-12 and about 0.5-2 for rodent tissues. The significance of the decreasing $^{239}Pu/^{238}Pu$ ratio from soils to vegetation to rodent tissues is not clear at this time. It may indicate that ^{238}Pu in the Trinity environs is more mobile than ^{239}Pu.

Table 2. The $^{239}Pu/^{238}Pu$ ratio in some ecosystem components collected in the fallout zone of the Trinity detonation.

	$^{239}Pu/^{238}Pu$		
Type Sample	$\bar{x}$	S.E.*	No. samples
Soils			
0-2.5 cm	19	2.2	9
2.5-7.5 cm	18	1.7	7
Remainder	9.0	2.3	6
Vegetation			
Grasses	12	2.0	13
Forbs	7.6	1.9	16
Shrubs	8.0	1.6	9
Rodents			
Liver	0.44	0.10	3
Lungs	1.0	0.28	8
[illegible]	1.8	0.29	16
Carcass	1.7	0.65	30

* S.E. = $\frac{\text{standard deviation}}{\sqrt{\text{number of samples}}}$

The Pu content of three samples of Trinitite from GZ measured 3.2 nCi/g, 1.5 nCi/g and 1.2 nCi/g with an average $^{239}Pu/^{238}Pu$ ratio of 21 $\pm$ 0.8 (1 σ). These Pu concentrations are about an order of magnitude higher than the Pu concentrations in GZ soils. The ^{241}Am concentrations of the Trinitite samples measured 0.5 nCi/g, 0.024 nCi/g and 0.033 nCi/g. Preliminary data from radioecological studies at Los Alamos[1] indicated that ^{241}Am may enter biological systems to a greater degree than Pu and, consequently, may be of equal or greater importance than Pu as a contaminant in natural [illegible] systems.

A wide variety of additional radionuclides were identified in Trinitite, including ^{133}Ba, 152, ^{155}Eu, ^{60}Co, ^{137}Cs and ^{90}Sr-^{90}Y.

The average gross gamma radiation measurements obtained outside the GZ area were not significantly different from the measurements obtained at remote locations or what could be considered to be the natural background radiation levels for the central New Mexico area.[7] Measured values of radiation also fall within the range of 12-20 μR/h suggested by Cowan[13] as being normal for the northern White Sands Missile Range elevation depending on the geological composition of the earth's crust. The measured values within GZ were significantly above background levels and approached a maximum of one mR/h under the measurement circumstances noted previously.

Results of this preliminary investigation indicated that the general pattern of Pu distribution in soils, vegetation, and rodents as a function of distance from GZ was similar to the findings of Leitch[5] and Olafson et al.[8] However, there has been an increased migration of Pu into the soils since the last measurements were made about 20 years ago. Concentrations of Pu in all sample types of the present study were generally similar to the findings of 20 years ago at Trinity Site. However, the limited number of samples analyzed in the present study does not allow an adequate comparison, and more refined studies are currently in progress.

Literature Cited

1. Hakonson, T. E., J. W. Nyhan, L. J. Johnson and K. V. Bostick. 1973. Ecological investigation of radioactive materials in waste discharge areas at Los Alamos. Los Alamos Scientific Laboratory Report LA-5282-MS.

2. Blair, W. F. 1943. Ecological distribution of mammals in the Tularosa Basin, New Mexico. Contr. Lab. Vert. Biol. Univ. of Michigan 20: 1-24.

3. Larson, K. H., J. L. Leitch, W. F. Dunn, J. W. Neel, J. H. Olafson, E. E. Held, J. Taylor, W. J. Cross, and A. W. Bellamy. 1951. Alpha activity due to the 1945 Atomic bomb detonation at Trinity, Alamogordo, New Mexico. Univ. of California Report UCLA-108.

4. Shields, Lora M. 1956. Zonation of vegetation within the Tularosa Basin, New Mexico. The Southwest Naturalist 1(2): 49-680.

5. Leitch, J. L. 1951. Summary of the radiological findings in animals from the biological surveys of 1947, 1948, 1949 and 1950. Univ. of California Report UCLA-111.

6. Herceg, J. E. 1972. Environmental monitoring in the vicinity of the Los Alamos Scientific Laboratory. Los Alamos Scientific Laboratory Report LA-4970.

7. Johnson, L. J. 1972. Los Alamos land areas environmental radiation, 1972. Los Alamos Scientific Laboratory Report LA-5097-MS.

8. Olafson, J. H., H. Nishita and K. H. Larson. 1957. The distribution of plutonium in the soils of central and northeastern New Mexico as a result of the atomic bomb test of July 16, 1945. Univ. of California Report UCLA-406.

9. Olafson, J. H. and K. H. Larson. 1961. Plutonium, its biology and environmental persistence. Univ. of California Report UCLA-501.

10. Francis, C. W. 1973. Plutonium mobility in soils and uptake in plants: a review. J. Environ. Quality, 2(1): 67-70.

11. Larson, K. H. 1958. Unpublished data. Cited in reference 9.

12. Whicker, F. W. 1973. Radioecology of some natural organisms and systems in Colorado. Eleventh Annual Progress Report on Atomic Energy Commission Contract AT(11-1)-1156.

13. Cowan, F. P. 1959. Natural radiation background. In Radiation Hygiene Handbook, McGraw-Hill Book Co., N.Y.